Reply to Anonymous Referee #1

We thank the reviewer for the careful reading of our manuscript and helpful comments. We have revised the manuscript following the suggestion, as described below.

General Comments

This manuscript presents the organic aerosol simulations in West China using WRF-Chem. Two types of SOA formation treatments are used, and the simulations are evaluated against monitoring station observations (O_3 and $PM_{2.5}$), campaign sampling site measurements (carbonaceous aerosols), and MODIS AOD retrievals (aerosol optical depth). The authors quantify the SOA fraction in OA and the contribution of different anthropogenic emission sources to OA at different environmental settings (urban, rural and background). The paper stands in a good form with some polishing.

Specific Comments

1) Pull together all measurement data and place in a separate sub-section in Section 2. This may make the structure flow more fluently. The measurement data should include those used for the model evaluation, such as from the monitoring stations, the field campaign sites, and the MODIS AOD retrievals. Instrumentation for the OA and EC filter field measurements should be described.

We have included a sub-section in Section 2 summarizing all the measurement data as follows:

"2.5 Measurement Data

The measurement data include temperature, relative humidity, and wind observations at the Jinghe meteorological station, hourly near-surface O_3 and $PM_{2.5}$ concentrations at ambient monitoring stations in Xi'an and surrounding areas, and daily filter measurements of $PM_{2.5}$, OC, and EC at 29 sites in GZB during the field campaign and at Institute of Earth Environment, Chinese Academy of Sciences (IEECAS) in Xi'an during the springtime from 2009 to 2013. The observation sites are categorized into three types regarding their locations: 18 urban sites, 10 rural sites, and 1 background site as shown in Figure 1b. The background site is located in the Qinling Mountains, which is far away from the residential areas. The daily filter samples are obtained on prefired (900 °C, 3 h) 47-mm Whatman QM-A quartz-fiber filters by mini-volume air samplers (Airmetrics, Eugene, OR) at 5 L min^{-1} flow rates.

The measured $PM_{2.5}$ and OC concentrations averaged over the 29 sites in GZB during the *study period are compared with the springtime* $PM_{2.5}$ *and OC observations at IEECAS site from 2009 to 2013 in Figure 3 along with the standard deviations. The springtime* $PM_{2.5}$ *concentrations at IEECAS site from 2009 to 2013 remain about 150* $\mu g\ m^{-3}$ *with small fluctuations, which is close to the* $PM_{2.5}$ *level during the study period. The springtime OC concentrations at IEECAS site from 2009 to 2013 vary from 14* $\mu g\ m^{-3}$ *to 22* $\mu g\ m^{-3}$*. The mean OC concentration during the study period is about 19* $\mu g\ m^{-3}$*, close to the medium level at IEECAS site from 2009 to 2013. Hence, the* $PM_{2.5}$ *and OC levels during the study period can well represent the springtime* $PM_{2.5}$ *and OC pollutions in GZB within recent years.*

The OC/EC ratio approach has been widely employed to evaluate the OA concentration from the filter measured OC and EC concentrations (Strader, 1999; Cao et al., 2003; 2004). The following method is used to derive the 'measured' POA, SOA, and OA mass concentrations from EC and OC filter measurements:

POC = EC × (POC/EC ratio)
SOC = OC – POC
POA = POC × (POA/POC ratio)
SOA = SOC × (SOA/SOC ratio)
OA = POA + SOA

Cao et al. (2007) have analyzed the OC and EC concentrations in 14 cities over China in 2003 and proposed primary OC/EC ratios for different cities in China during winter and summer (Table 3). Numerous studies have been performed to investigate the POA/POC and SOA/SOC ratios (Aiken et al., 2008; Yu et al., 2009; Yu, 2011), which can be used to obtain OA concentrations from measured EC and OC concentrations. In this study, the POC/EC, POA/POC, SOA/SOC ratios are assumed to be 2.4, 1.2, and 1.6, respectively, based on the previous studies (Cao et al., 2007; Aiken et al., 2008; Yu et al., 2009; Yu, 2011). It is worth to note that, these assumed values might affect the model-measurement comparisons.

Using the measured EC and OC concentrations, the SOA/EC ratio is calculated:

(SOA/EC ratio) = (SOA/SOC ratio) × (SOC/EC ratio)
= (SOA/SOC ratio) × [(OC – POC) / EC]
= (SOA/SOC ratio) ×[(OC/EC ratio) – (POC/EC ratio)]

And the POA/EC ratio is derived as follows:

(POA/EC ratio) = (POA/POC ratio) × (POC/EC ratio)"

MODIS AOD retrievals have been removed from the manuscript according to the suggestion of Anonymous Reviewer #2.

2) Add a table summarizing the source-categorized anthropogenic emission in Xi'an and its surroundings in Section 2. This may facilitate the discussions on the OA source apportionment in Section 3.2.4.

We have included a table and a short description in the last paragraph of Section 2.3 about the anthropogenic emissions in Xi'an and surrounding areas: "*Table 2 presents the primary organic* *carbon and volatile organic compounds emissions from anthropogenic sources in Xi'an and surrounding areas in April.*"

Table 2 Primary organic carbon and volatile organic compounds emissions from anthropogenic sources in Xi'an and surrounding areas (the area surrounded by the white line in Figure 1c) in April

Anthropogenic Emission Sectors	*Primary Organic Carbon (Mg)*	*Volatile Organic Compounds (Mg)*
Industry	*2486.0*	*7634.5*
Power Plant	*0.0*	*81.7*
Residential	*4988.8*	*1704.4*
Transportation	*318.5*	*736.9*

3) Clarify Section 3.2. It is not clear to me why certain values of OC/EC and OA/OC ratios are used and how "measured" POA and SOA values are derived in this study. To derive the POA and SOA concentrations from the OA and EC measurements, you need to have the values for the POC/EC, SOC/EC, POA/POC, and SOA/SOC, but in the description there is no presumed value for the SOC/EC ratio. Also please justify the uses of the values of POC/EC (2.4), POA/POC (1.2), and SOA/SOC (1.6), and you may also address that these values may affect the model-measurement comparisons. In addition, it would be interesting to compare the assumed POA/EC and SOA/EC ratios with the simulated counterparts.

We have clarified in Section 2.5:

" *The OC/EC ratio approach has been widely employed to evaluate the OA concentration from the filter measured OC and EC concentrations (Strader, 1999; Cao et al., 2003; 2004). The following method is used to derive the 'measured' POA, SOA, and OA mass concentrations from EC and OC filter measurements:*

$$POC = EC \times (POC/EC\ ratio)$$
$$SOC = OC - POC$$
$$POA = POC \times (POA/POC\ ratio)$$
$$SOA = SOC \times (SOA/SOC\ ratio)$$
$$OA = POA + SOA$$

Cao et al. (2007) have analyzed the OC and EC concentrations in 14 cities over China in 2003 and proposed primary OC/EC ratios for different cities in China during winter and summer (Table 3). Numerous studies have been performed to investigate the POA/POC and SOA/SOC ratios (Aiken et al., 2008; Yu et al., 2009; Yu, 2011), which can be used to obtain OA concentrations from measured EC and OC concentrations. In this study, the POC/EC, POA/POC, *SOA/SOC ratios are assumed to be 2.4, 1.2, and 1.6, respectively, based on the previous studies (Cao et al., 2007; Aiken et al., 2008; Yu et al., 2009; Yu, 2011). It is worth to note that these assumed values might affect the model-measurement comparisons.*

Using the measured EC and OC concentrations, the SOA/EC ratio is calculated:
(SOA/EC ratio) = (SOA/SOC ratio) × (SOC/EC ratio)
= (SOA/SOC ratio) × [(OC – POC) / EC]
= (SOA/SOC ratio) ×[(OC/EC ratio) – (POC/EC ratio)]
And the POA/EC ratio is derived as follows:
(POA/EC ratio) = (POA/POC ratio) × (POC/EC ratio)"

We have compared the "measured" POA/EC and SOA/EC ratios with simulations in the second paragraph of Section 3.2.1: "*The simulated POA/EC and SOA/EC ratios using the NT-SOA module are 3.23 and 2.23, close to the 'measured' 2.88 and 2.58, respectively. However, the ratios using the T2-SOA module are 6.54 and 0.22, respectively, dramatically deviated from the 'measurements'.*"

4) In the end of Page 15, you rightly point out that "*future studies need to be performed to further improve SOA simulations and OA source apportionment*". There are many factors contributing to the OA, SOA in particular, simulation uncertainties, including measurements, meteorology, emissions, and SOA formation mechanisms and treatments; even right modeling results might be due to wrong reasons (e.g., right concentrations but wrong O/C ratios). Elaborating a little bit more on what aspects of the SOA modeling can be improved would be insightful.

We have elaborated the improvements on SOA modeling in further studies in the last paragraph of Section 4: "*It is worth to note that many factors contribute to the OA and SOA simulation uncertainties, such as measurements, meteorology, emissions, SOA formation mechanisms and treatments et al.; even right modeling results might be caused by wrong reasons (e.g., right concentrations but wrong O/C ratios). To better simulate the SOA formation, the SOA mechanisms and treatments need further revising and improving to reasonably represent OA formation and development in the atmosphere, such as including the oxidation degree, rather than only nine surrogates categorized by saturation concentrations. Additionally, reducing uncertainties from meteorological fields simulations and emissions are also imperative to improve the SOA simulation. In addition to OA, other aerosol species, such as sulfate, nitrate, ammonium, and mineral dust, also play important roles in the haze formation. Further source appointment of those aerosol species is needed to support the design of mitigation strategies.*"

5) In Section 3.2.4 and Table 2, you may add the OA/$PM_{2.5}$ fractions, which may provide more

scientific information for devising the haze control strategy, since OA is only one important component of the haze in the GZB.

We have included the OA/$PM_{2.5}$ fractions in Table 5 and added the following sentence in the second paragraph of Section 3.2.3: "*Table 5 presents OA concentrations and contributions and OA contributions to $PM_{2.5}$ from anthropogenic emissions averaged over the simulation period at urban, rural, and background sites.*"

6) P3, lines 65-67. For the upper limit fractions, SOA/$PM_{2.5}$ has a higher value (77%) than SOA/OA does (71%)? Specify the investigation region in the Sun et al. (2012) study.

We have corrected the sentence in Section 1: "*Huang et al. (2014) have reported that OA constitutes a major fraction (30~50%) of the total $PM_{2.5}$ in Beijing, Shanghai, Guangzhou, and Xi'an during severe haze pollution events (Table 1), and SOA contributed 44~71% of OA mass concentrations.*"

We have specified the investigation region in the Sun et al. (2012) in Section 1: "*Using the ACSM (Aerosol Chemical Speciation Monitor) measurements analyzed by PMF (positive matrix factorization), Sun et al. (2012) have showed that the oxygenated organic aerosols (OOA, a surrogate of SOA) dominate OA composition in Beijing, with a contribution of 64% on average from 26 June to 28 August 2011.*"

7) Line 85, start a new paragraph from "*During the period from 20. . .*".

We have started a new paragraph from "*During the period from 20. . .*" in the last but one paragraph of Section 1.

8) Ls 200-202, the morning elevation could also be contributed by the morning rush hour emissions. What do the $PM_{2.5}$ diurnal profiles look like?

We have revised that sentence in the last paragraph of Section 3.1.1 as "*The $PM_{2.5}$ mass concentrations are generally elevated in the morning during the episode, probably contributed by the weak or calm horizontal winds, slow development of PBL, and the morning rush hour emissions. The $PM_{2.5}$ levels fall down in the afternoon, caused by the enhanced dispersion in the horizontal and vertical directions.*"

9) Ls 224-232, describe how AOD is estimated in the model.

The AOD estimation follows the method in Li et al. (ACP, 2011), in which the extinction efficiency, SSA, and asymmetry factor are calculated using the Mie theory at a given wavelength.

The aerosol spectrum is first divided into 48 bins from 0.002 μm to 2.5 μm, with radius r_i. When the bin's radius is less than 0.1 μm, the interval of bins ranges from 0.001 to 0.005 μm. When the bin's radius is greater than 0.1 μm, the interval is increased to 0.025 to 0.25 μm. The aerosols are classified into four types: (1) internally mixed sulfate, nitrate, ammonium, hydrophilic organics, hydrophilic black carbon, and water; (2) hydrophobic organics; (3) hydrophobic black carbon; and (4) other unidentified aerosols. These four kinds of aerosols are assumed to be mixed externally. The aerosol optical thickness (AOT or τ_a) at a given wavelength in a given atmospheric layer k is determined by the summation over all types of aerosols and all bins:

$$\tau_{\mathrm{a}}(\lambda,k)=\sum_{i=1}^{48}\sum_{j=1}^{4}Q_{\mathrm{e}}(\lambda,r_i,j,k)\pi r_i^2 n(r_i,j,k)\Delta z_k$$

where $n(r_i, j, k)$ is the number concentration of j-th kind of aerosols in i-th bin. Δz_k is the depth of an atmospheric layer.

Following the suggestion from Anonymous Reviewer #2, we have excluded the AOD comparison in this study.

10) Ls 254-257, conflicting. The difference between 33.6% in T2-SOA and 4.3% in NT-SOA is not "not remarkable".

We have revised that sentence in the first paragraph of Section 3.2.1 as "*The T2-SOA and NT-SOA modules exhibit differently in simulating OA in GZB, as shown in Figure 10a.*"

11) Ls 286-303, Discussions on the SOA/OA fractions in the two paragraphs overlap, and you can merge them.

The paragraphs discuss the SOA fractions (Figure 13) and SOA pathways (Figure 14), respectively. We have moved the following sentence to Section 2.5: "*The observation sites are categorized into three types regarding their locations: 18 urban sites, 10 rural sites, and 1 background site as shown in Figure 1b. The background site is located in the Qinling Mountains, which is far away from the residential areas.*"

12) Ls 331-333, the emission source is one reason; another one can be due to the rapid transport and transformation processes.

We have updated that sentence in the last paragraph of Section 3.2.3: "*It is worth to note that, the OA contributions from residential, transportation, and industry emissions are comparable at the* *urban and rural sites, which probably is due to the rapid transport and transformation process between urban and rural areas. In addition, the urbanization and industrialization in GZB may also rapidly diminish the OA source difference between the rural and urban areas.*"

13) Ls 341-342, Model results are compared not only against O_3 and $PM_{2.5}$ from the monitoring stations, but also against the $PM_{2.5}$ carbonaceous components from the OC and EC field filter measurements, and against the MODIS AOD.

We have clarified in Section 4: "*Model results are compared with the meteorological observations, hourly* O_3 *and* $PM_{2.5}$ *measurements in Xi'an and surrounding areas, and the* $PM_{2.5}$ *carbonaceous components from the OC and EC field filter measurements in GZB.*"

14) Page 24 Figure 1 caption, describe the black "circle" (urban borderline?).

We have added in Figure 1 caption: "*The black circle in (b) and (c) shows the ring expressway of Xi'an.*"

Technical comments

1) Line 175, pollution

We have changed "pollutions" to "pollution" in the first paragraph of Section 3.1.1.

2) Line 183, concentrations substantially increase to

We have changed sentence in the first paragraph of Section 3.1.1: "*The observed and simulated* O_3 *mass concentrations substantially increase to more than 80* $\mu g\ m^{-3}$ *at 15:00 BJT with the enhancement of photochemical activities.*"

3) Line 195, delete well

We have deleted "well" in that sentence in the second paragraph of Section 3.1.2.

4) Line 198, the air quality with respect to $PM_{2.5}$

We have changed "*the air quality in Xi'an ...*" to "*the air quality with respect to* $PM_{2.5}$ *in Xi'an ...*" in the second paragraph of Section 3.1.1.

5) L275-276, change to something like "Since the NT-SOA module significantly improves the POA and SOA simulations, we will use the NT-SOA OA simulations for further comparisons

and the OA source apportioning. Figure 11. . .”

We have changed the sentence in the first paragraph of Section 3.2.2 as “*The NT-SOA module significantly improves POA and SOA simulations, so we use the NT-SOA OA simulations for further comparisons and OA source apportionment. Figure 12 displays the spatial distributions of OA, POA, and SOA simulated by the NT-SOA module against the measurement in GZB.*”

6) 310, verify to estimate

We have changed “verify” to “estimate” in the first paragraph of Section 3.2.3.

7) Line 324, emissions are

We have changed “emission is” to “emissions are” in the second paragraph of Section 3.2.3.